INVESTIGATIONS OF ATOMIC AND NUCLEAR ENERGY LEVELS BY CRYSTAL DIFFRACTION AND PHOTO ELECTRON SPECTROSCOPY

BY

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ALMQVIST & WIKSELLS BOKTRYCKERI AB

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Inaugural Dissertation

BY

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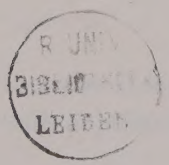

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to Margareta, Ingrid and Staffan

". . . what is the use of a book," thought Alice
"without pictures or conversations?"

LEWIS CARROLL

This thesis consists of studies of *atomic energy levels* by *X-ray absorption-*, *emission-*, and *photo electron*-spectroscopy. The crystal diffraction technique is further extended for the study of *nuclear levels*.

Our knowledge of inner atomic levels is based for the most part on X-ray data obtained in the 1920's and 1930's. The increasing requirements of modern physics have made renewed investigations with improved X-ray techniques necessary, and stimulated interest in the application of precision electron spectroscopy to the study of electronic shell structure. A few examples will be mentioned. At the latest conference of the International Union of Crystallography in Stockholm in 1959, which was mainly devoted to the topics of X-ray wavelength problems and precision lattice parameter determinations, the need for accurate X-ray emission line studies was strongly emphasized. Furthermore, in precision beta-ray spectroscopy, X-ray data are demanded for calibration purposes as well as for the interpretation of conversion electron spectra in terms of nuclear transitions. The insufficient accuracy of the available values of atomic level energies has in many cases placed a limitation on the precision attained. The fruitful study of mesic atoms also demands accurate knowledge of the structure of the normal atom. In the important field of measurements of fundamental constants in physics, an attempt is now being made at this Institute to solve the familiar problem of the "h/e discrepancy" with the use of a method proposed by Kai Siegbahn. Again, the accuracy of the determination is to a large extent dependent on the precision of available X-ray emission line data.

The present investigation contributes to precision X-ray and atomic level energy data on K and L levels in 24 heavier elements. Most of these, such as mercury and the rare earths, have not been studied previously with comparable accuracy. The advantageous combination of the methods of crystal diffraction and photo electron spectroscopy is employed here for the first time. The photo electron method is superior to X-ray absorption spectroscopy as regards determinations of electron binding energies in several respects. Measurements of X-ray emission lines are, however, a valuable complement to the photo electron data, which is demonstrated in Papers II, V, and VI. Systematic comparisons of the results of photo electron determinations with those obtained from X-ray absorption and emission spectra are also found to furnish interesting material to the current discussion of electronic band structure.

Recently, high precision X-ray measurements of the fine structure in heavy elements have caused a great deal of interest from another aspect, as they have brought out nuclear size- and quantum electrodynamical effects on the atomic levels. The interpretation of the experimental data requires theoretical expressions for the spin

doublet splitting, which are discussed in Paper V in the light of the present results. It appears to the author that previous conclusions require revision.

Finally it should be pointed out that the clean and direct diffraction techniques of X-ray spectroscopy can be successfully applied to investigations of nuclear radiation. The diffraction spectrometer has a capacity for precision energy and intensity measurements, demonstrated in Paper VII, which can now be fully utilized, owing to the high thermal neutron fluxes available for source activation and for the study of (n, γ) reactions from in-pile sources.

The thesis consists of the following papers, a summary of which is given below.

I. Olof Beckman, Pär Bergvall and Bertil Axelsson: A precision curved crystal X-ray and gamma-ray spectrometer, Arkiv för Fysik *14*, 419 (1958).

II. Olof Beckman, Bertil Axelsson and Pär Bergvall: Effect of spectrometer window on the K absorption edge of Au, Arkiv för Fysik *15*, 567 (1959).

III. Olof Beckman and Pär Bergvall: Precision measurement of K X-ray spectrum of Hg, The Physical Review *114*, 1280 (1959).

IV. Pär Bergvall: Precision measurement of $K\alpha$ X-ray lines from rare earth elements, Arkiv för Fysik *16*, 57 (1959).

V. Pär Bergvall and Stig Hagström: Atomic level energies in rare earth elements, Arkiv för Fysik *17*, 61 (1960).

VI. Pär Bergvall, Olle Hörnfeldt and Carl Nordling: Crystal diffraction and photo electron measurements of K and L levels in the elements 47 Ag to 52 Te, Arkiv för Fysik *17*, 113 (1960).

VII. Pär Bergvall: Precision measurements of gamma energies and intensities by crystal diffraction, Arkiv för Fysik *17*, 125 (1960).

The design of a curved crystal diffraction spectrometer according to new principles, which permit great simplicity in the mechanical construction is described in Paper I. A discussion of the systematic errors in the wavelength determinations is given, and the instrument is tested by measuring certain well established X-ray lines.

Paper II describes measurements of the K absorption discontinuity in gold. The "thickness effect", which implies a systematic shift of the edge, dependent on absorber thickness and caused by influence of the finite spectrometer window, is observed in transmission for the first time. Theoretical estimates of the size of the effect are in accordance with experiment. The extrapolated energy of the K limit shows agreement, within experimental errors, with energies obtained by addition of the $K\alpha_1$, α_2 emission lines, measured separately, to the L_{III}, L_{II} levels according to photo electron data reported by Sokolowski.

Wavelength values of all the electric dipole and quadrupole lines, in the K spectrum of mercury are reported in Paper III, which is based on previous work by Beckman and present measurements of the four strongest lines. The accuracy is better than that of previous data by a factor of 10. The same applies to the measurements of the $K\alpha$ lines in the rare earth elements, reported in Paper IV.

In Paper V photo electron measurements of the three L levels in the rare earths

are reported. Good agreement is found with the X-ray emission line data in Paper IV regarding the L_{II}–L_{III} level difference, and K level energies, obtained by addition of the emission lines to the levels, are given. The theoretical expressions for the spin doublet splitting and the possibility of obtaining a quantitative estimate of nuclear size effects are discussed.

Combined X-ray emission and photo electron data on K and L levels are given in Paper VI. Measurements are made on six elements from silver to tellurium, an energy region where both methods yield optimal accuracy, which for the K emission line determinations means better than 10 parts per million.

The extension of the diffraction spectrometer measurements up to energies of 1.2 MeV is described in Paper VII, and the accuracy of the energy determinations of gamma lines is demonstrated. With the use of data from inter alia Papers IV and V, the energies of six standard reference conversion electron lines in different energy regions are given. An efficiency calibration is reported, and the problems of relative intensity measurements are discussed.

ACKNOWLEDGEMENTS

This research has been carried out at the Institute of Physics of the University of Uppsala. I owe a debt of gratitude to Professor Kai Siegbahn for his generous support and never-failing interest in this investigation. It is also a pleasure to acknowledge the kind interest and encouragement of Professor Per Ohlin.

I was introduced to the experimental techniques of X-ray spectroscopy in 1954 by Dr Olof Beckman, who also initiated the construction of and measurements with the diffraction spectrometer. His stimulating guidance is deeply appreciated.

In 1956, I was given the opportunity of visiting Professor Jesse W. M. Du Mond at the California Institute of Technology in Pasadena, where I was able to gain experience and establish contacts which have since been of great value to me. I am particularly grateful to Professor Du Mond for maintaining a most personal interest in my work.

Among my friends and colleagues at the Institute to whom I am indebted for their support, I especially wish to thank Dr Carl Nordling for giving freely of his time to discussions of my problems.

Uppsala in March, 1960. *Pär Bergvall*